Final Progress Report

Program: NASA NRA-97-MTPE-16

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Title Validation of the lidar retrieval of aerosol extinction

Period June, 1999 - May 2002

Part I: Overview

Summary:

The goal of this project is to facilitate the integration of Light Detection and Ranging (lidar) measurements in building a global aerosol climatology. Lidar is sensitive to aerosol even at modest concentrations, provides vertical resolution, and is capable of surveying large regions of the troposphere - indeed, upcoming satellite missions will provide truly global surveys. However, quantitative interpretation of lidar data requires knowledge of the extinction-to-backscatter ratio, S_a . As such, the focus of our project has been to develop a database and an approach to constraining S_a . As shown below, knowledge of S_a for tropospheric aerosols was highly unconstrained at the outset of this project and is much better constrained today (due to the efforts of several groups, not just our own). Two case studies using spaceborne lidar data are used below to demonstrate the vast reduction of uncertainty in lidar retrievals that is associated with moving from unconstrained S_a to constrained S_a .

Prior to this project, our group developed a method for calibrated, in situ measurement of S_a (Doherty et al., 1999, Appl. Optics, 38, 1823). During the course of this project, we deployed this technology in several field campaigns to obtain characteristic values for a wide range of aerosol types and to study the physical/chemical/thermodynamic parameters that control variations in S_a . This grant was the primary source of support for one field campaign (LINC) and for the analysis of S_a from all field campaigns.

While considerable progress has been made over the three years of this project, a great deal remains to be done. As documented below, the recent increase in data on S_a has been extraordinary. These data come from our approach as well as approaches based on Raman lidar, slant-path lidar, and sunphotometry. The challenge ahead is to integrate these data and to develop and test methods of constraining S_a throughout the global troposphere. The planned launch in 2004 of the ESSP3-CENA satellite (a lidar satellite to fly in formation with the many aerosol sensors on NASA's Aqua satellite) provides a compelling target date for achieving this larger goal. As described herein, our research has helped to lay the foundation for this larger project, which will form the basis of a subsequent proposal.

Yearly breakdown of activities

year 1: June 1999 - May 2000 INDOEX, LINC*, and SEAS field campaigns

year 2: June 2000 - May 2001 ACE-Asia field campaign year 3: June 2001 - May 2002 analysis, publication

*LINC campaign was primarily funded through this grant.

Part II: Publications

Primary publications for GACP bibliography:

- Anderson, T. (1999) Chapter 5: Extinction-to-backscatter ratio database in <u>PICASSO-CENA: Lidar Algorithm Theoretical Basis Document White Papers</u>, NASA Langley, Oct. 8, 1999. 55-63.
 - URL: http://www.atmos.washington.edu/~cheeka/PC/whitepapers.html
- Anderson, T. L., Masonis, S. J., Covert, D. S., Charlson, R. J. and Rood, M. J., (2000): In-situ measurement of the aerosol extinction-to-backscatter ratio at a polluted, continental site, *J. Geophys. Res.*, 105, 26907-26915.
 - URL: http://www.atmos.washington.edu/~cheeka/linc/SaLINCjgr.html
- Masonis, S. J., Franke, K., Ansmann, A., Mueller, D., Althausen, D., Ogren, J. A., Jefferson, A. and Sheridan, P. J., (2001): An intercomparison of aerosol light extinction and 180° backscatter as derived using in-situ instruments and Raman lidar during the INDOEX field campaign, *J. Geophys. Res.*, in press. URL: not yet available
- Stephens, G. L., Vaughan, M., Engelen, R. J. and Anderson, T. L., (2001): Toward retrieving properties of the tenuous atmosphere using space-based Lidar measurements, *J. Geophys. Res.*, in press. URL: not yet available

Additional publications partially supported by this project:

- Anderson, T. L., Covert, D. S. and Ghan, S. J., (2001): A practical index of aerosol type for transport modeling, satellite retrievals, and in-situ measurements, manuscript in preparation.
 - URL: http://www.atmos.washington.edu/~cheeka/Rmass/Rmass.html
- Anderson, T., Eisele, F., Schmidt, U., Tyndall, G. and Winker, D. (2001) Chapter 5: Instrumentation in <u>Atmospheric Chemistry in a Changing World</u>, G. Brasseur (Eds.) URL: not yet available
- Rodhe, H., Charlson, R. J. and Anderson, T. L., (2000): Avoiding circular logic in climate modeling, *Climatic Change*, 44, 419-422. URL: none

Part III: Detailed report

This section describes progress made toward an empirically based constraint on the aerosol extinction-to-backscatter ratio, S_a , for application to simple, elastic-scatter lidar systems, especially spaceborne lidar.

Definitions

Lidar equation: An elastic backscatter lidar responds to a convolution of local 180° backscattering, β , and path-integrated extinction, σ :

$$P(r) = \frac{C\beta(r)}{r^2} \exp \left[-2\int_0^r \sigma(r') dr' \right]$$
 (1)

where r is the range, P(r) is the detected power, and C is the instrument calibration constant.

Aerosol extinction-to-backscatter ratio, S_a : The deconvolution of β and σ can be accomplished given independent knowledge of the extinction-to-backscatter ratio, S. Since the optical properties of air are well known, the problem reduces to understanding the *aerosol* extinction-to-backscatter ratio, S_a .

$$S_a = \frac{\sigma_e}{\beta} = \frac{\sigma_s + \sigma_a}{\beta} \tag{2}$$

where σ_e is the extinction coefficient due to aerosol particles and σ_s , and σ_a are the components of aerosol extinction due to scattering and absorption, respectively.

Importance of S_a for quantitative backscatter and extinction retrievals

Sensitivity analysis has been performed using spaceborne lidar data from the LITE shuttle mission (Winker et al., 1996) under two alternate assumptions:

Unconstrained Sa

factor of 4 uncertainty (20-80 sr)

Constrained Sa

factor of 1.5 uncertainty (either 20-30 sr or 40-60 sr)

Results (shown graphically in Figures 1 and 2) can be summarized as follows:

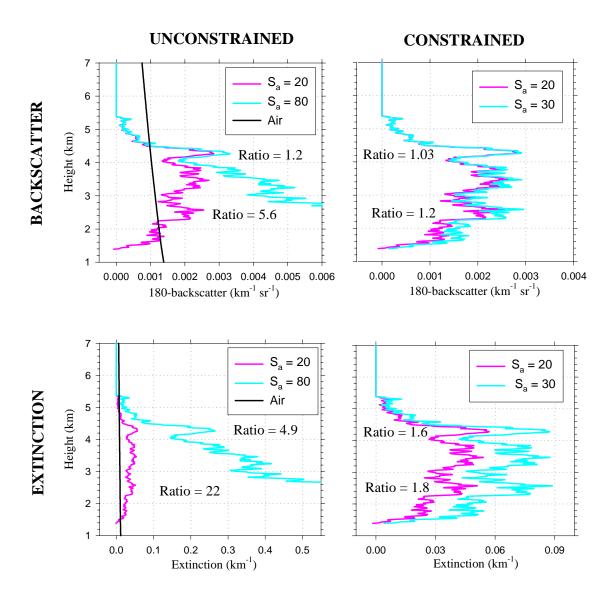
With unconstrained S_a you (only) get:

➤ accurate backscatter at top of first layer

With constrained S_a you get:

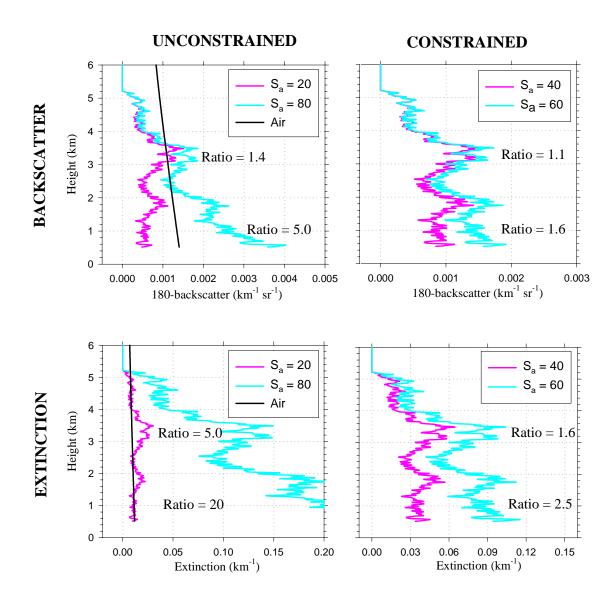
► accurate backscatter profiles

>considerable bounding of extinction profiles



data from Mark Vaughan, plot by Tad Anderson, July 1, 2001

<u>Figure 1:</u> Retrieved profiles of aerosol backscatter and extinction from spaceborne lidar acquired during the Lidar In-Space Technology (LITE) experiment, Sept., 1994. The "Ratio" values shown on the plots are the ratios of uncontrained to contrained values at the indicated location of the profile. The case shown applies to a Saharan dust layer over the Atlantic Ocean.



data from Mark Vaughan, plot by Tad Anderson, July 1, 2001

Figure 2: As in Fig. 1 except for a boundary layer aerosol beneath thin cirrus.

Empirical knowledge of Sa

Given its importance to lidar retrievals, we undertook the task of assessing the state of knowledge of S_a for tropospheric aerosols. Our initial literature review, in 1999, showed an overall paucity of measurements, but did permit three useful insights:

- 1. geographical location by itself does not constrain S_a
- 2. the measured range is about 10 to 100 sr
- 3. coarse-mode aerosol is generally toward low end of range; pollution aerosol is generally toward high end of range

The literature review is summarized in Table 1, below, adapted from Anderson et al. (2000).

Table 1: Measurements of S_a for tropospheric aerosols, as of 1999

Method	Location	$N^{(a)}$	S (sr)
slant-path lidar (b)	mixed layer, west U.S. (Tucson, AZ)	57	15-82
horizontal lidar (c)	Netherlands	10	10-50
horizontal lidar (d)	marine surface layer, Australian coast	10	40-80
multi-wave lidar (e)	marine boundary layer (tropical Atlantic)	1	<30
	mixed layer over rainforest (S. Amer.)	1	43-60
	Saharan dust aloft (tropical Atlantic)	2	15-62
backscatter-sonde (f)	rural, arid SW U.S.	12	42
	rural western U.S. lower troposphere	4	15-30
	rural western U.S. upper troposphere	4	15-60
space-borne lidar (g)	smoke layers in SH upper troposphere	7	50-90
Raman lidar (h)	polluted boundary layer over Leipzig,	1	55-95
	Germany		

^a Approximate number of independent samples.

But it is not enough to have individual measurements of S_a that are not coupled to other aerosol and atmospheric properties. In order to (i) determine the variability of S_a , and (ii) establish parameterizations of S_a and (iii) understand the accuracy of these parameterizations, we need:

b Determines effective S over mixed layer at 694 nm. (Spinhirne et al., 1980; Reagan et al., 1984; 1988)

c 1064 nm laser. No information on location of instrument or type of aerosol investigated. (de Leeuw et al., 1986)

d 532 nm laser beam aimed 2 m over ocean surface from coastal site. Effect of waves not assessed. (Young et al., 1993).

 $^{^{}e}$ S at 600 nm is constrained by Mie calculations based on the measured wavelength variation of β_{p} . (Sasano and Browell, 1989)

^f S reported at 690 nm. Method requires significant wavelength and angular adjustments, based on assumed size distributions and Mie calculations. (Rosen et al., 1997a, b)

^g S at 532 nm is constrained to values yielding physically plausible lidar retrievals under the assumption of constant S throughout upper troposphere. (Kent et al., 1998)

h S at 532 nm determined from independent extinction measurements using nitrogen-Raman. (Ansmann et al., 1992; Muller et al., 1998)

- Sufficiently large data sets for statistical analysis
- imultaneous, collocated measurements of thermodynamic state
- >simultaneous, collocated measurements of aerosol composition
- >vertical resolution sufficient to distinguish major aerosol layers

Viewed in this light, the situation just two years ago with regard to empirical measurements of S_a looked bleak, as shown in Table 2a:

Table 2a: S_a measurements with significant sample sizes before **1999**

method	location	notes	S _a range	•	dynam.		cal
slant lidar	Tucson, AZ	a	15 - 82	57	yes	no	no

[&]quot;N": estimated number of independent samples

However, spurred by the prospect of global lidar data from satellites, the community has made great strides in developing adequate data sets. Table 2b, below, shows data sets currently published or in press. Keep in mind that several other data sets (e.g. from PRIDE, SAFARI, ACE-Asia, AERONET) are in the works. This Table reflects analysis by our group and is adapted from a co-authored paper (Stephens et al., 2001).

[&]quot;Sa range": range that encompasses 95% of the data

⁽a) Lidar measurements at several slant angles determines effective S over mixed layer at 694 nm; sporadic daily samples over 3 years [Reagan et al., 1988].

Table 2b: S_a measurements with significant sample sizes

method	location	notes	S_a	sample	thermo-	aerosol	verti-
			range	size	dynam.	compo-	cal
					state?	sition?	resol.?
slant lidar	Tucson, AZ	a	15 - 82	57	yes	no	no
nephelometry	central Illinois	b	27 - 75	95	yes	yes	no
nephelometry	Indian Ocean	c	33 - 91	168	yes	yes	yes
nephelometry	Hawaiian coast	d	26 - 33	35	yes	yes	no
lidar/sunpho.	Indian Ocean	e	25 - 75	102	yes	surface	no
Raman lidar	Oklahoma	f	45 - 87	2556	yes	no	yes
nephelometer	Asian outflow	g	35 - 65	~200	yes	yes	yes

[&]quot;N": estimated number of independent samples

- (d) Method as in (b); continuous surface measurements over 8 days during onshore (marine) flow; N assumes 1 independent sample every 4 hours [Masonis, 2001].
- (e) Vertically pointing Micro Pulse Lidar and sunphotometer determine effective S over aerosol-laden lower troposphere; continuous data from ship during same experiment as in (c); N assumes one independent sample every 4 hours for the 17 cloud-free days of data [Welton et al., 2001].
- (f) Raman lidar determines local S from 0.8 km to about 4 km at 355 nm; continuous data from surface station over 2 years; N assumes 4 independent samples in vertical obtained every 4 hours over 213 days with 50% data loss due to cloud contamination [Ferrare et al., 2001].
- (g) Method as in (b); airborne data (30 to 6000 m altitude) from 19 flights over 6 weeks primarily in the Yellow Sea and Sea of Japan during the ACE-Asia campaign, Spring, 2000; N assumes 1 independent sample every 1 km vertical or 100 km horizont [Anderson, TL, Masonis, SJ, unpublished data].

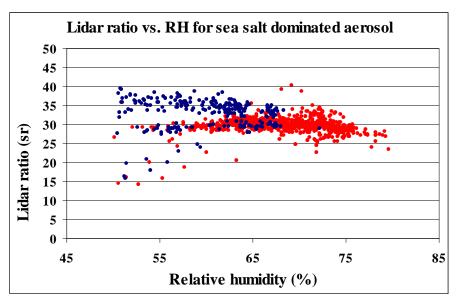
[&]quot;S_a range": range that encompasses 95% of the data

⁽a) Lidar measurements at several slant angles determines effective S over mixed layer at 694 nm; sporadic daily samples over 3 years [Reagan et al., 1988].

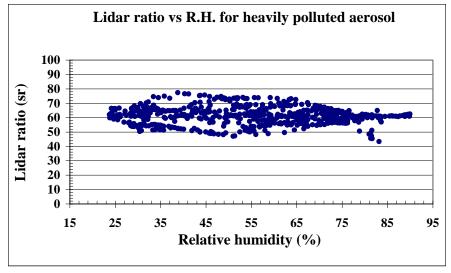
⁽b) Nephelometric method determines local S at 532 nm; continuous surface measurements over 5 weeks; N assumes 1 independent sample every 4 hours [Anderson et al., 2000].

⁽c) Method as in (b); airborne data (30 to 3500 m altitude) from 18 flights over 6 weeks during low-level offshore flow from surrounding continents; N assumes 1 independent sample every 1 km vertical or 100 km horizontal [Masonis, 2001; Masonis et al., 2001].

Two of these data sets reveal remarkable consistencies in the cases of clean marine seasalt (Figure 3) and sulfate-dominated pollution from the midwestern US (Figure 4).

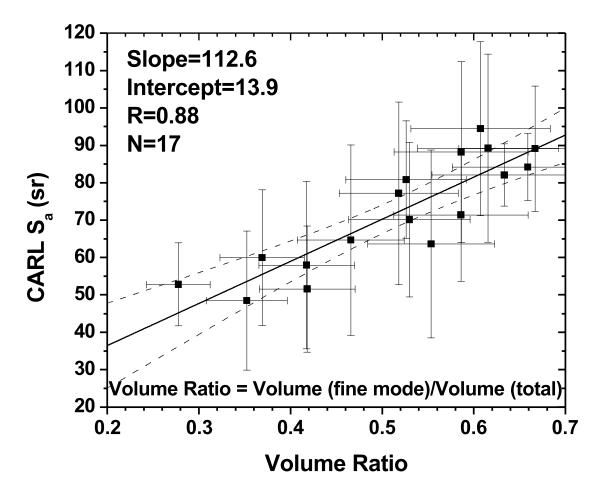


<u>Figure 3:</u> Lidar ratio versus the relative humidity at which it was measured during the SEAS field campaign, Bellows Fields, Oahu, Hawaii (March-April, 2000; Masonis, 2001, PhD dissertation, University of Washington). For most of the campaign the aerosol was characterized as clean marine (red), but some of the samples were influence by volcanic aerosol from Japan (blue).



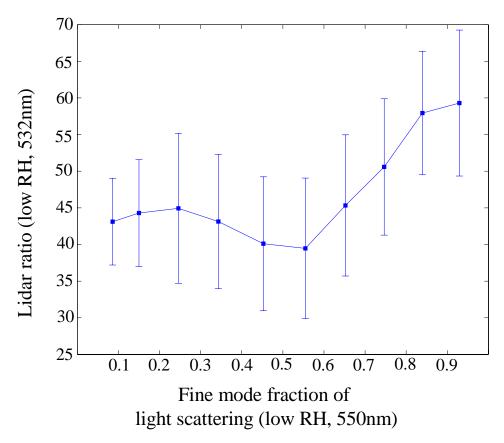
<u>Figure 4:</u> Lidar ratio versus the relative humidity at which it was measured. Nephelometric technique [Doherty et al., 1999, Appl. Optics, 38, 1823-1832.] These data were collected during the LINC field campaign in Bondville, Illinois (August-September, 1999; Anderson et al., 2000, J. Geophys. Res., 105, 26907-26915) during times when the aerosol was characterized as heavily polluted.

In other cases, the data reveal useful relationships for building parameterizations. For example, Fig. 5 shows a strong relation between S_a and the fraction of aerosol volume in fine mode.



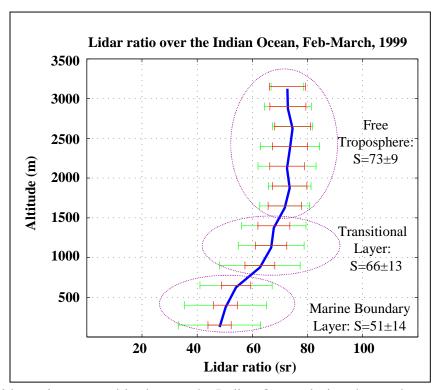
<u>Figure 5:</u> Aerosol extinction-to-backscatter ratio, S_a , at 355 nm wavelength measured by Raman lidar versus the fine-mode fraction of aerosol volume derived by inversion of sunphotometer/radiometer measurements. All measurements at DOE Southern Great Plains site in central Oklahoma. Cases chosen for comparison had aerosol optical depth (at 440 nm) greater than 0.2 and vertical variation in S_a less than 10 sr. [Ferrare et al., 2001, J. Geophys. Res., in press]

Similarly, Fig. 6 shows a relationship between S_a and the fraction of aerosol scattering in fine mode.

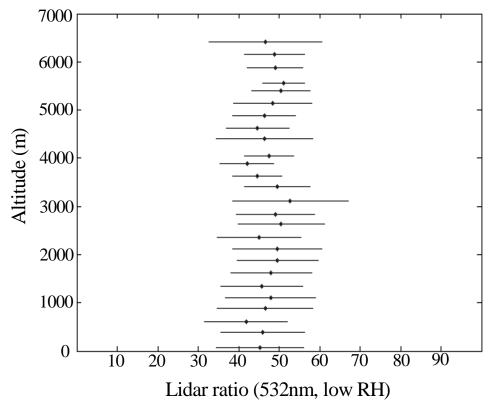


<u>Figure 6:</u> Airborne, nephelometric measurements of S_a versus the fine-mode fraction of aerosol light scattering, σ_{sp} . Data from 19 flights over the Yellow Sea and Sea of Japan during the Asian dust season, ACE-Asia. Means and standard deviations are shown. Curiously, the expected relationship was found only for fine-mode fractions of 50% or more. [Anderson, TL and Masonis, SJ, unpublished data]

Figures 7 and 8 show relationships, from two different campaigns, between S_a and height. For pollution aerosol advected over the Indian Ocean (INDOEX campaign), variation in the vertical has been shown to have a consistent structure (Fig. 7). For dust and pollution aerosol advected over the Yellow Sea and Sea of Japan (ACE-Asia), no systematic variation in the vertical was observed (Fig. 8).



<u>Figure 7:</u> Lidar ratio versus altitude over the Indian Ocean during the northeast monsoon (blue), as measured during INDOEX. The standard deviation at each altitude is shown in green, and the uncertainty of the mean (95% C.I.) in red.



<u>Figure 8:</u> Airborne, nephelometric measurements of S_a from 19 flights over the Yellow Sea and Sea of Japan during the Asian dust season, ACE-Asia. Means and standard deviations are shown for data grouped by altitude. [Anderson, TL and Masonis, SJ, unpublished data]

Dust enigma

There is conflicting evidence concerning the appropriate value of Sa for mineral dust aerosol. This section outlines the evidence on both sides.

Evidence that dust- S_a is low (20-30 sr at 532 nm):

<u>1. "theory":</u> 20 +/- 5 sr

OPAC "mineral aerosol model" combined with Mie calculations [Ackermann, 1998]. However, calculation assumes spheres.

2. Tucson measurements:

~25 sr

Extensive slant-path lidar measurements by Reagan et al. [1988] at 694 nm show a dominant cluster at $S_a = \sim 25$ sr. However, *some* measurements, even with measured mass dominated by coarse mode, do show values around 40 sr.

3. LITE measurements:

25 - 35 sr

Lofted aerosol layers were observed by LITE over Atlantic during a large Saharan dust outflow event. Retrieval of optical depth and S_a by transmission method indicated S_a = 25 - 35 sr at 532 nm [Karyampudi et al., 1999]. However, estimate uses a correction for multiple scattering which is based on an assumed scattering phase function.

Evidence that dust- S_a is high (35-50 sr at 532 nm):

1. Non-spherical theory:

S_a should be higher

For coarse-mode, non-spherical particles, spherical (Mie) calculations significantly overestimate 180° backscattering but have little effect on total scattering [Mishchenko et al., 1995] such that they <u>underestimate extinction-to-backscatter</u>. However, these non-spherical predictions for 180° backscattering have not been confirmed experimentally.

2. Sahara dust measurements in ACE-2:

~ 36 sr

Welton et al. [2000] Powell et al. [2000] 37 +/- 9 sr

35 +/- 5 sr

However, this is only one dust event, and it is not known how much non-dust (e.g. pollution) aerosol was mixed with the dust.

3. Dust measurements in ACE-Asia:

43 +/- 6 sr

Nephelometric technique; data from flights/dust events; dust cases identified when coarse-mode fraction of scattering is > 80% [Anderson and Masonis, unpublished data.] However, results are preliminary and unpublished.

Further caveats:

- 1. These discrepancies could reflect real atmospheric variability. It is possible that dust- S_a is not a constant, but varies as a function of:
 - * source region (via changes in mineral composition)
 - * distance from source (via changes in size-distribution)
- 2. Measurements and analysis aimed at resolving this discrepancy (e.g. via method comparisons) are underway.

Conclusions and Implications

▶ Profiles of aerosol backscatter and extinction from elastic backscatter lidar (e.g. as will be deployed from space in upcoming NASA missions) can be retrieved with greater accuracy if S_a can be constrained. Sensitivity studies indicate that knowing S_a to within about a factor of 1.5 is required for meaningful, quantitative retrievals.

Geographical location does not, by itself, provide an adequate constraint.

 \triangleright Empirical knowledge of S_a has grown tremendously in the past 2 years. These data reveal many circumstances where S_a is either invariant or varies in a predictable way.

The appropriate value(s) of S_a for mineral dust is uncertain and is an area of active research.

 \triangleright Creating a global look-up table of S_a for use by satellite lidar appears to be feasible.

To make optimal use of the growing empirical database, the look-up table must include all of the following elements:

- geographical location, vertical location, season
- major aerosol types expected in that time/location
- * criteria for deciding which of the expected aerosol types is present. These criteria should not require knowledge of S_a. Likely candidates would be top-of-layer backscatter magnitude for detecting plumes and the backscatter color ratio as an indicator of coarse-mode fraction.

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